Spin Injection and Detection in a Mesoscopic Superconductor at Low Temperatures

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We theoretically study nonequilibrium spin transport in a superconducting wire connected by tunnel junctions to two ferromagnetic metal wires, each of which serves as an injector or detector of spin-polarized electron current. We present a set of Boltzmann equations to determine nonequilibrium quasiparticle distributions in this system, and obtain an analytical expression for the nonlocal spin signal in the case of small injection current. It is shown that the quasiparticle distribution in the ferromagnetic metal for detection strongly affects the magnitude of the spin signal. At low temperatures, since nonequilibrium quasiparticles created by the tunneling from the superconductor dominate thermally excited ones, the spin signal becomes independent of temperature. This explains the convergence of the spin signal with decreasing temperature observed in a recent experiment by Poli *et al.*

KEYWORDS: spin accumulation, spin imbalance, energy imbalance, spin signal

1. Introduction

Experimental studies on spin injection and detection in a normal metal have attracted considerable attention recently in the field of spintronics.[1–5] More than two decades ago, Johnson and Silsbee[1] performed the first experiment on this subject by using a large normal metal sample with two electrodes made of a ferromagnetic metal, where each electrode serves as a spin injector or detector. Spin-polarized electrons created near the injector diffuse in the normal metal, and spin imbalance is transmitted to the detector if spin-flip scattering does not suppress it. They found an evidence of spin imbalance by measuring an open-circuit voltage induced at the detector. Several experiments using devices in the mesoscopic regime have been reported to date.[2–4] The most popular device consists of a thin normal metal wire connected to a few ferromagnetic metal wires. In this system, we supply injection current $I_{\rm inj}$ with spin polarization $P_{\rm spin}$ into the normal metal from one of ferromagnetic metals and measure an open-circuit voltage between another ferromagnetic metal and the normal metal. Let $V_{\rm p}$ ($V_{\rm ap}$) be the open-circuit voltage when the magnetizations of the two ferromagnetic metals are parallel (antiparallel). We are interested in the nonlocal spin signal defined by

$$R_{\rm spin} = \frac{V_{\rm p} - V_{\rm ap}}{I_{\rm inj}}, \tag{1}$$

which crucially depends on the spin diffusion length $\lambda_{\rm sf}$ and the distance d between the injection and detection points. In the case where the normal metal and the two ferromagnetic metals are connected by tunnel junctions, the spin signal is given by[3,5]

$$R_{\rm spin} = P_{\rm spin}^2 R_{\rm N} {\rm e}^{-\frac{d}{\lambda_{\rm sf}}}, \tag{2}$$

where $R_{\rm N} \equiv \rho_{\rm N}\lambda_{\rm sf}/A_{\rm N}$ with $\rho_{\rm N}$ and $A_{\rm N}$ being the resistivity and the cross-sectional area of the normal metal, respectively.

Spin injection and detection in a superconductor is also attracted considerable attention.[5–11] Our primary interest focuses on how the spin signal is modified by the transition to the superconducting state. Takahashi and Maekawa[5] studied this problem and predicted that

$$R_{\rm spin} = \frac{1}{2f_0(\Delta)} P_{\rm spin}^2 R_{\rm N} {\rm e}^{-\frac{d}{\lambda_{\rm sf}}}, \tag{3}$$

where $f_0(\Delta) = 1/(\exp(\Delta/T)+1)$ with the superconducting energy gap Δ and temperature T. This indicates that $R_{\rm spin}$ exponentially increases with decreasing T. They claimed that this modification is caused by the increase of spin resistivity due to the opening of the energy gap Δ. The increase of $R_{\rm spin}$ with decreasing T has been successfully observed in the recent experiment by Poli *et al.*[11] However, there remain a few points to be clarified. We focus on the following two points. Firstly, Takahashi and Maekawa implicitly assume in their derivation of eq. (3) that spin imbalance in a superconductor can be described by a shift of spin-dependent chemical potential. This assumption cannot be justified at low temperatures, where energy relaxation due to phonon scattering is not strong. Secondly, Poli *et al.* observed convergence of $R_{\rm spin}$ with decreasing temperature. This behavior cannot be explained by eq. (3).

In this paper, we theoretically study nonequilibrium spin transport in a hybrid system consisting of a superconducting wire and two ferromagnetic metal wires. Each ferromagnetic metal is connected by a tunnel junction to the superconductor, and serves as an injector or detector of spin-polarized quasiparticles. We present a set of Boltzmann equations governing nonequilibrium quasiparticles in this system. We focus on the case of small injection current at low temperatures, and obtain not only the quasiparticle distribution in the superconducting wire but also that in the ferromagnetic metal wire for detection. On the basis of the resulting nonequilibrium distributions, we derive an analytical expression for the nonlocal spin signal. It is shown that although the spin signal originates from spin imbalance transmitted to the detection junction, its magnitude is not solely determined by the spin imbalance but is strongly affected by

the quasiparticle distribution in the ferromagnetic metal. We observe that when T is higher than a crossover temperature T_{cross}, the spin signal exponentially increases with decreasing T reflecting the reduction of thermally excited quasiparticles in the ferromagnetic metal. At low temperatures below T_{cross}, however, the magnitude of the spin signal is determined by nonequilibrium quasiparticles created by the tunneling from the superconductor instead of thermally excited ones, and the spin signal becomes independent of T. This explains the convergence of the spin signal with decreasing T observed by Poli *et al.*[11]

In the next section, we present a set of Boltzmann equations to describe nonequilibrium quasiparticle distributions in the hybrid system consisting of a superconducting wire and two ferromagnetic metal wires. In §3, we obtain nonequilibrium quasiparticle distributions in this system by solving the set of Boltzmann equations, and derive an analytical expression of the spin signal on the basis of the resulting quasiparticle distributions. In §4, we compare our theoretical result with the recent experimental result. We set $\hbar = k_{\mathrm{B}} = 1$ throughout this paper.

2. Formulation

Let us consider the hybrid system consisting of a superconducting wire and two ferromagnetic metal wires (see Fig. 1). We assume that the superconductor is connected by a tunnel junction to each ferromagnetic metal. The left and right junctions serve as spin injector and detector, respectively. We adopt a simple one-dimensional model for this device assuming that the superconductor and the ferromagnetic metals are very thin. We introduce the x axis in the superconductor on which the left and right junctions are located at $x = x_{\mathrm{inj}}$ and $x = x_{\mathrm{det}}$, respectively, and the y axis in the left (right) ferromagnetic metal on which the injection (detection) junction is located at $y = y_{\mathrm{inj}}$ (y_{det}). We denote by d the separation between the two junctions. That is, $d \equiv x_{\mathrm{det}} - x_{\mathrm{inj}}$. We inject spin-polarized current into the superconductor by applying a bias voltage V_{inj} across the injection junction, and measure an induced open-circuit voltage V_{det} across the detection junction under the condition that net current flow vanishes between the superconductor and the ferromagnetic metal for detection. We simply assume that the spin polarization P_{spin} of the injection current is proportional to the difference between the density of states $N_{\mathrm{F}\uparrow}$ for up-spin electrons and $N_{\mathrm{F}\downarrow}$ for down-spin electrons. The spin polarization is expressed as

$$P_{\mathrm{spin}} = \frac{N_{\mathrm{F}\uparrow} - N_{\mathrm{F}\downarrow}}{N_{\mathrm{F}\uparrow} + N_{\mathrm{F}\downarrow}}. \tag{4}$$

We assume that spin relaxation in the superconductor is caused by spin-flip scattering due to spin-orbit interaction as well as magnetic impurities.

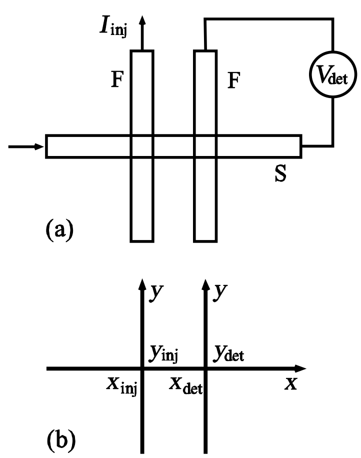

Fig. 1. (a) Schematic picture of the model system consisting of a superconducting wire (S) and two ferromagnetic metal wires (F). The left (right) ferromagnetic metal serves as an injector (detector) of spin-polarized electron current. (b) Spatial coordinates used in the text.

To present an expression for the tunneling current across each junction, we consider nonequilibrium quasiparticle distributions in the superconductor and the ferromagnetic metals. We first introduce the quasiparticle distribution function $g_{\mathrm{FL}\sigma}$ in the left ferromagnetic metal for injection, where $\sigma =\uparrow, \downarrow$ is the spin variable. We assume $g_{\mathrm{FL}\sigma}(y, \epsilon) = f_0(\epsilon - eV_{\mathrm{inj}})$ with the Fermi-Dirac distribution function $f_0(\epsilon)$. Here and hereafter, we measure quasiparticle energy from the chemical potential of the superconductor not only in the superconductor but also in the ferromagnetic metals. We next introduce the quasiparticle distribution function $g_{\mathrm{S}\sigma}$ in the superconductor. In terms of four distribution functions $f_{\mathrm{L}+}$, $f_{\mathrm{L}-}$, $f_{\mathrm{T}+}$ and $f_{\mathrm{T}-}$ for nonequilibrium quasiparticles, we express it as[12–16]

$$g_{\mathrm{S}\uparrow}(x, \epsilon) = f_0(\epsilon) + f_{\mathrm{L}+}(x, \epsilon) + f_{\mathrm{L}-}(x, \epsilon) + f_{\mathrm{T}+}(x, \epsilon) + f_{\mathrm{T}-}(x, \epsilon), \tag{5}$$

$$g_{\mathrm{S}\downarrow}(x, \epsilon) = f_0(\epsilon) - f_{\mathrm{L}+}(x, \epsilon) + f_{\mathrm{L}-}(x, \epsilon) + f_{\mathrm{T}+}(x, \epsilon) - f_{\mathrm{T}-}(x, \epsilon). \tag{6}$$

The four distribution functions satisfy

$$f_{\mathrm{L,T}+}(x, -\epsilon) = f_{\mathrm{L,T}+}(x, \epsilon), \tag{7}$$

$$f_{\mathrm{L,T}-}(x, -\epsilon) = -f_{\mathrm{L,T}-}(x, \epsilon). \tag{8}$$

Note that $f_{\mathrm{L}+}$ describes spin imbalance, while $f_{\mathrm{T}+}$ describes charge imbalance.[17,18] The other two functions $f_{\mathrm{L}-}$ and $f_{\mathrm{T}-}$ describe total energy imbalance and energy imbalance between up-spin and down-spin quasiparticles, respectively. Finally, we introduce the distribution function $g_{\mathrm{FR}\sigma}$ in the right ferromagnetic metal in which nonequilibrium quasiparticles appear due to quasiparticle tunneling from the superconductor. We express it as

$$g_{\mathrm{FR}\sigma}(y, \epsilon) = f_0(\epsilon - eV_{\mathrm{det}}) + f_{\mathrm{F}\sigma}(y, \epsilon). \tag{9}$$

We hereafter assume that the magnitude of the energy gap Δ is unaffected by spin injection everywhere in the superconductor. This allows us to consider $f_{\mathrm{L}\pm}(x, \epsilon)$ and $f_{\mathrm{T}\pm}(x, \epsilon)$ only for $|\epsilon| > \Delta$. The nonequilibrium distribution functions $f_{\mathrm{L}\pm}$, $f_{\mathrm{T}\pm}$ and $f_{\mathrm{F}\sigma}$ are governed by Boltzmann equations which we present below.

The tunneling current at the injection junction is given by

$$I_{\rm inj}(V_{\rm inj}) = \frac{\Delta}{eR_{\rm inj}} J_1(V_{\rm inj}, T), \tag{10}$$

where $R_{\rm inj}$ is the tunnel resistance of the injection junction and

$$J_1(V,T) = \frac{1}{\Delta}\int_0^\infty {\rm d}\epsilon N_1(\epsilon)\bigl(f_0(\epsilon - eV) - f_0(\epsilon + eV)\bigr) \tag{11}$$

with N_1 being the normalized density of states in the superconductor, given by $N_1(\epsilon) = |\epsilon|/\sqrt{\epsilon^2 - \Delta^2}$ for $|\epsilon| > \Delta$ in the BCS limit. In deriving eq. (10), we have ignored small contributions arising from nonequilibrium quasiparticles in the superconductor. The tunneling current between the superconductor and the right ferromagnetic metal for detection is expressed as

$$I_{\rm det}(V_{\rm det}) = I_{\rm q}(V_{\rm det}) + I_{\rm F}(V_{\rm det}) - I_{\rm S}(V_{\rm det}), \tag{12}$$

where $I_{\rm q}$ is the ordinary tunneling current arising from thermally excited quasiparticles, while $I_{\rm F}$ represents the contribution from nonequilibrium quasiparticles induced in the ferromagnetic metal. The third term $I_{\rm S}$ represents the contribution from spin and charge imbalances. They are expressed as

$$I_{\rm q}(V_{\rm det}) = \frac{\Delta}{eR_{\rm det}} J_1(V_{\rm det}, T), \tag{13}$$

$$\begin{aligned} I_{\rm F}(V_{\rm det}) = {}& \frac{1}{eR_{\rm det}}\int_0^\infty {\rm d}\epsilon\, N_1(\epsilon) \\ & \times \Biggl(\frac{1+P_{\rm spin}}{2}\bigl(f_{\rm F\uparrow}(y_{\rm det},\epsilon) + f_{\rm F\uparrow}(y_{\rm det},-\epsilon)\bigr) \\ & + \frac{1-P_{\rm spin}}{2}\bigl(f_{\rm F\downarrow}(y_{\rm det},\epsilon) + f_{\rm F\downarrow}(y_{\rm det},-\epsilon)\bigr)\Biggr), \end{aligned} \tag{14}$$

$$\begin{aligned} I_{\rm S}(V_{\rm det}) = {}& \frac{2}{eR_{\rm det}}\int_0^\infty {\rm d}\epsilon\, N_1(\epsilon) \\ & \times \bigl(P_{\rm spin} f_{\rm L+}(x_{\rm det},\epsilon) + f_{\rm T+}(x_{\rm det},\epsilon)\bigr), \end{aligned} \tag{15}$$

where $R_{\rm det}$ is the tunnel resistance of the detection junction. In eq. (15), the first term with $f_{\rm L+}$ represents the contribution from spin imbalance and is the origin of the spin signal, while the second term with $f_{\rm T+}$ represents that from charge imbalance. In deriving eqs. (14) and (15), we have assumed the parallel alignment of magnetizations. The corresponding expressions for the antiparallel alignment is obtained by reversing the sign of $P_{\rm spin}$.

To present Boltzmann equations for $f_{\rm L\pm}$ and $f_{\rm T\pm}$, we introduce the Usadel equation[19] for the quasiclassical retarded Green's functions g^R and f^R,

$${\rm i}\epsilon f^R(\epsilon) + \Delta g^R(\epsilon) - \frac{1}{\tau_{\rm m}} g^R(\epsilon) f^R(\epsilon) = 0, \tag{16}$$

where $\tau_{\rm m}$ represents the magnetic impurity scattering time and we have assumed that the superconductor is spatially homogeneous. The spectral functions N_1, N_2, R_1 and R_2 are defined as

$$g^R(\epsilon) = N_1(\epsilon) + {\rm i}R_1(\epsilon), \tag{17}$$

$$f^R(\epsilon) = N_2(\epsilon) + {\rm i}R_2(\epsilon). \tag{18}$$

In terms of the spectral functions, the Boltzmann equations are expressed as[12–16]

$$\begin{aligned} & D_{\rm S}\left(N_1^2(\epsilon) - R_2^2(\epsilon)\right)\partial_x^2 f_{\rm L+}(x,\epsilon) \\ & \quad - \frac{4}{3\tau_{\rm so}}\left(N_1^2(\epsilon) - R_2^2(\epsilon)\right) f_{\rm L+}(x,\epsilon) \\ & \quad - \frac{4}{3\tau_{\rm m}}\left(N_1^2(\epsilon) + R_2^2(\epsilon)\right) f_{\rm L+}(x,\epsilon) \\ & \quad + P_{\rm L+}(x,\epsilon) = 0, \end{aligned} \tag{19}$$

$$D_{\rm S}\left(N_1^2(\epsilon) - R_2^2(\epsilon)\right)\partial_x^2 f_{\rm L-}(x,\epsilon) + P_{\rm L-}(x,\epsilon) = 0, \tag{20}$$

$$\begin{aligned} & D_{\rm S}\left(N_1^2(\epsilon) + N_2^2(\epsilon)\right)\partial_x^2 f_{\rm T+}(x,\epsilon) \\ & \quad - \frac{1}{\tau_{\rm conv}(\epsilon)} f_{\rm T+}(x,\epsilon) + P_{\rm T+}(x,\epsilon) = 0, \end{aligned} \tag{21}$$

$$\begin{aligned} & D_{\rm S}\left(N_1^2(\epsilon) + N_2^2(\epsilon)\right)\partial_x^2 f_{\rm T-}(x,\epsilon) \\ & \quad - \frac{4}{3\tau_{\rm so}}\left(N_1^2(\epsilon) + N_2^2(\epsilon)\right) f_{\rm T-}(x,\epsilon) \\ & \quad - \frac{4}{3\tau_{\rm m}}\left(N_1^2(\epsilon) - N_2^2(\epsilon)\right) f_{\rm T-}(x,\epsilon) \\ & \quad - \frac{1}{\tau_{\rm conv}(\epsilon)} f_{\rm T-}(x,\epsilon) + P_{\rm T-}(x,\epsilon) = 0, \end{aligned} \tag{22}$$

where $D_{\rm S}$ is the diffusion constant, $\tau_{\rm so}$ and $\tau_{\rm conv}$ are the spin-orbit scattering time and the charge imbalance conversion time, respectively, and $P_{\rm L\pm}$ and $P_{\rm T\pm}$ are the injection terms which represent quasiparticle tunneling between the superconductor and the left ferromagnetic metal. The injection terms are given as[16,20]

$$\begin{aligned} P_{\rm L+}(x,\epsilon) = {}& \frac{\delta(x - x_{\rm inj}) N_1(\epsilon)}{4e^2 N_{\rm S} A_{\rm S} R_{\rm inj}} \\ & \times \Bigl[P_{\rm spin}\bigl(f_0(\epsilon - eV_{\rm inj}) - f_0(\epsilon + eV_{\rm inj})\bigr) \\ & - 2\bigl(f_{\rm L+}(x_{\rm inj},\epsilon) + P_{\rm spin} f_{\rm T+}(x_{\rm inj},\epsilon)\bigr)\Bigr], \end{aligned} \tag{23}$$

$$\begin{aligned} P_{\rm L-}(x,\epsilon) = {}& \frac{\delta(x - x_{\rm inj}) N_1(\epsilon)}{4e^2 N_{\rm S} A_{\rm S} R_{\rm inj}} \\ & \times \Bigl[f_0(\epsilon + eV_{\rm inj}) + f_0(\epsilon - eV_{\rm inj}) - 2f_0(\epsilon) \\ & - 2\bigl(f_{\rm L-}(x_{\rm inj},\epsilon) + P_{\rm spin} f_{\rm T-}(x_{\rm inj},\epsilon)\bigr)\Bigr], \end{aligned} \tag{24}$$

$$\begin{aligned} P_{\rm T+}(x,\epsilon) = {}& \frac{\delta(x - x_{\rm inj}) N_1(\epsilon)}{4e^2 N_{\rm S} A_{\rm S} R_{\rm inj}} \\ & \times \Bigl[f_0(\epsilon - eV_{\rm inj}) - f_0(\epsilon + eV_{\rm inj}) \\ & - 2\bigl(P_{\rm spin} f_{\rm L+}(x_{\rm inj},\epsilon) + f_{\rm T+}(x_{\rm inj},\epsilon)\bigr)\Bigr], \end{aligned} \tag{25}$$

$$\begin{aligned} P_{\rm T-}(x,\epsilon) = {}& \frac{\delta(x - x_{\rm inj}) N_1(x,\epsilon)}{4e^2 N_{\rm S} A_{\rm S} R_{\rm inj}} \\ & \times \Bigl[P_{\rm spin}\bigl(f_0(\epsilon + eV_{\rm inj}) + f_0(\epsilon - eV_{\rm inj}) - 2f_0(\epsilon)\bigr) \\ & - 2\bigl(P_{\rm spin} f_{\rm L-}(x_{\rm inj},\epsilon) + f_{\rm T-}(x_{\rm inj},\epsilon)\bigr)\Bigr], \end{aligned} \tag{26}$$

where $N_{\rm S}$ and $A_{\rm S}$ are the density of states at the Fermi level in the normal state and the cross-sectional area of the superconducting wire, respectively. We can ignore

$f_{L\pm}(x_{inj}, \epsilon)$ and $f_{T\pm}(x_{inj}, \epsilon)$ in these injection terms when the injection current is small. It should be noted that inelastic phonon scattering has been ignored in the Boltzmann equations because its role is not relevant at low temperatures, in which we are interested. We have also ignored very small contributions to $f_{L\pm}$ and $f_{T\pm}$ arising from the coupling with the right ferromagnetic metal for detection.

We turn to quasiparticle distributions in the right ferromagnetic metal for detection. We note that in obtaining I_F, the spin-dependence of $f_{F\sigma}$ is not important as long as the spin polarization is small. This indicates that we need not consider complicated spin-dependent dynamics of nonequilibrium quasiparticles. We thus define

$$f_{F+}(y, \epsilon) = \frac{f_{F\uparrow}(y, \epsilon) + f_{F\downarrow}(y, \epsilon)}{2}, \tag{27}$$

and approximate the expression of I_F as

$$I_F(V_{det}) = \frac{1}{eR_{det}} \int_0^\infty d\epsilon\, N_1(\epsilon) \times \big(f_{F+}(y_{det}, \epsilon) + f_{F+}(y_{det}, -\epsilon)\big). \tag{28}$$

We present an appropriate Boltzmann equation for f_{F+}. We ignore roles of spin-flip scattering since the spin-dependence is not important for our argument. However, we must consider the energy relaxation process due to phonon scattering. The reason for this is as follows. Since quasiparticles in the ferromagnetic metal are induced by the tunneling from the superconductor with the energy gap Δ, their excitation energy is of the order of Δ and no quasiparticle is directly created in the subgap region. Quasiparticles in such a nonequilibrium situation inevitably experience the energy relaxation. We thus assume that f_{F+} obeys

$$D_F \partial_y^2 f_{F+}(y, \epsilon) - \frac{1}{\tau_e(\epsilon - eV_{det})} f_{F+}(y, \epsilon) + P_{F+}(y, \epsilon) = 0, \tag{29}$$

where D_F is the diffusion constant averaged over spin directions and τ_e is the energy relaxation time with $\epsilon - eV_{det}$ being the quasiparticle energy measured from the chemical potential of the ferromagnetic metal. The source term P_{F+} describing quasiparticle tunneling from the superconductor is given by

$$P_{F+}(y, \epsilon) = \frac{\delta(y - y_{det}) N_1(\epsilon)}{2e^2 N_F A_F R_{det}} \big(f_0(\epsilon) - f_0(\epsilon - eV_{det}) - f_{F+}(y_{det}, \epsilon) + f_{L-}(x_{det}, \epsilon) + f_{T+}(x_{det}, \epsilon)\big), \tag{30}$$

where $N_F \equiv (N_{F\uparrow} + N_{F\downarrow})/2$ and A_F is the cross-sectional area of the ferromagnetic metal. For the expression of τ_e, we adopt

$$\frac{1}{\tau_e(\epsilon)} = 2 \int_{-\infty}^{\infty} d\epsilon' \sigma_F(\epsilon, \epsilon') \times \left(\coth\left(\frac{\epsilon' - \epsilon}{2T}\right) - \tanh\left(\frac{\epsilon'}{2T}\right)\right) \tag{31}$$

with

$$\sigma_F(\epsilon, \epsilon') = \frac{\alpha_F}{4} \mathrm{sign}(\epsilon' - \epsilon) \times (\epsilon' - \epsilon)^2, \tag{32}$$

where α_F characterizes the strength of electron-phonon coupling. For $|\epsilon| \gg T$, we approximately obtain

$$\frac{1}{\tau_e(\epsilon)} = \frac{\alpha_F}{3} |\epsilon|^3. \tag{33}$$

3. Spin Signal

In this section, we solve the Boltzmann equations and obtain the spin signal defined in eq. (1) by evaluating V_p and V_{ap}. Note that V_p (V_{ap}) is the open-circuit voltage induced across the detection junction when the magnetizations of the injector and detector are in the parallel (antiparallel) alignment. We determine V_p and V_{ap} by the condition of $I_{det} = 0$. We assume that the magnitude of V_p and V_{ap} is much smaller than Δ/e. However, we do not assume $V_{inj} \ll \Delta/e$. We focus on the case where the injection current is so small that injected quasiparticles are populated only near the gap edge (i.e., $|\epsilon| \approx \Delta$). In this case, f_{T+} and f_{T-} quickly relaxes because the conversion time becomes very short near the gap edge.[12,18,21,22] Therefore, we ignore f_{T+} and f_{T-} in the following argument. Furthermore, the smallness of the injection current also allows us to ignore $f_{L+}(x_{inj}, \epsilon)$ and $f_{L-}(x_{inj}, \epsilon)$ in the injection terms given in eqs. (23) and (24).

We first assume that magnetic impurities are absent (i.e., $\tau_m^{-1} = 0$) and define the spin-flip scattering time τ_{sf} as

$$\frac{1}{\tau_{sf}} = \frac{4}{3\tau_{so}}. \tag{34}$$

In this case, the spectral functions for $|\epsilon| > \Delta$ are simply given by

$$N_1(\epsilon) = \frac{|\epsilon|}{\sqrt{\epsilon^2 - \Delta^2}}, \tag{35}$$

$$R_2(\epsilon) = \frac{\mathrm{sign}(\epsilon)\Delta}{\sqrt{\epsilon^2 - \Delta^2}}, \tag{36}$$

and $N_2(\epsilon) = R_1(\epsilon) = 0$. This indicates that $N_1^2(\epsilon) - R_2^2(\epsilon) = 1$. We first obtain $f_{L+}(x_{det}, \epsilon)$ by solving eq. (19). Note that $f_{L+}(x, \epsilon)$ decays exponentially as a function of $|x - x_{inj}|$ and this decay is characterized by the spin-diffusion length given by $\lambda_{sf} = \sqrt{D_S \tau_{sf}}$. We obtain

$$f_{L+}(x_{det}, \epsilon) = \frac{P_{spin} \lambda_{sf}}{8e^2 N_S A_S R_{inj} D_S} \Sigma_+(\epsilon, V_{inj}) e^{-\frac{d}{\lambda_{sf}}} \tag{37}$$

with

$$\Sigma_+(\epsilon, V_{inj}) = N_1(\epsilon)\big(f_0(\epsilon - eV_{inj}) - f_0(\epsilon + eV_{inj})\big). \tag{38}$$

Next, we obtain $f_{L-}(x_{det}, \epsilon)$ which is necessary to obtain $f_{F+}(y_{det}, \epsilon)$. A special care must be paid in solving eq. (20) since no relaxation process is included in this equation. The relaxation of f_{L-} is mainly caused by the phonon-mediated recombination process, which is described by adding the following nonlinear term[20]

$$I_{L-}(x, \epsilon) = -4 \int d\epsilon' \sigma_S(\epsilon, \epsilon') \times \left(N_1(\epsilon) N_1(\epsilon') - R_2(\epsilon) R_2(\epsilon')\right) f_{L-}(x, \epsilon) f_{L-}(x, \epsilon') \tag{39}$$

to eq. (20). Here, $\sigma_{\rm S}(\epsilon,\epsilon')$ is identical to $\sigma_{\rm F}(\epsilon,\epsilon')$ in eq. (32) if $\alpha_{\rm F}$ is replaced by $\alpha_{\rm S}$. From this expression, we observe that the corresponding decay length $L_{\rm c}$ becomes very long when the injection current is small and therefore $|f_{\rm L-}(x,\epsilon)| \ll 1$. We thus assume that $L_{\rm c}$ is longer than, or at least of the order of, the length of the superconducting wire, and adopt the boundary condition that $f_{\rm L-}$ vanishes at each end of the superconducting wire. We further assume that the distance between the injection junction and each end of the superconductor is nearly equal to L, and $L \gg d$. Under this assumption, we approximately obtain

$$f_{\rm L-}(x_{\rm det},\epsilon) = \frac{L}{8e^2 N_{\rm S} A_{\rm S} R_{\rm inj} D_{\rm S}} \Sigma_-(\epsilon, V_{\rm inj}) \tag{40}$$

with

$$\Sigma_-(\epsilon, V_{\rm inj}) = N_1(\epsilon)\big(f_0(\epsilon - eV_{\rm inj}) + f_0(\epsilon + eV_{\rm inj}) - 2f_0(\epsilon)\big). \tag{41}$$

If $L \gg L_{\rm c}$, we must replace L in eq. (40) with $L_{\rm c}$. Finally, we obtain $f_{\rm F+}(y_{\rm det},\epsilon)$. It should be emphasized that $I_{\rm F}$ containing $f_{\rm F+}$ becomes relevant in the low temperature regime where the ordinary contribution $I_{\rm q}$ is exponentially suppressed due to the opening of the energy gap. In this regime, the term with $f_0(\epsilon) - f_0(\epsilon - eV_{\rm det})$ in $P_{\rm F+}$ can be neglected. Furthermore, since $f_{\rm T+}$ can be ignored, the dominant contribution to $P_{\rm F+}$ arises from the term with $f_{\rm L-}$. This indicates that nonequilibrium quasiparticles are created by energy imbalance in the superconductor. We thus approximate $P_{\rm F+}$ as

$$P_{\rm F+}(y,\epsilon) = \frac{\delta(y - y_{\rm det}) N_1(\epsilon)}{2e^2 N_{\rm F} A_{\rm F} R_{\rm det}} f_{\rm L-}(x_{\rm det},\epsilon). \tag{42}$$

Solving eq. (29), we obtain

$$f_{\rm F+}(y_{\rm det},\epsilon) = \frac{\lambda_{\rm e}(\epsilon - eV_{\rm det})}{4e^2 N_{\rm F} A_{\rm F} R_{\rm det} D_{\rm F}} N_1(\epsilon) f_{\rm L-}(x_{\rm det},\epsilon), \tag{43}$$

where the energy relaxation length $\lambda_{\rm e}$ is given by $\lambda_{\rm e}(\epsilon) = \sqrt{D_{\rm F}\tau_{\rm e}(\epsilon)}$. Combining this and eq. (40) and noting that quasiparticles are populated near the gap edge (i.e., $|\epsilon| \approx \Delta$), we approximately obtain

$$\begin{aligned} &f_{\rm F+}(y_{\rm det},\epsilon) + f_{\rm F+}(y_{\rm det},-\epsilon) \\ &= \frac{L}{8e^2 N_{\rm S} A_{\rm S} R_{\rm inj} D_{\rm S}} \frac{\lambda_{\rm e}(\Delta)}{4e^2 N_{\rm F} A_{\rm F} R_{\rm det} D_{\rm F}} \\ &\quad \times N_1(\epsilon)\Sigma_-(\epsilon, V_{\rm inj}) \frac{3eV_{\rm det}}{\Delta}. \end{aligned} \tag{44}$$

From eqs. (28) and (44), we observe that $I_{\rm F} = 0$ at $V_{\rm det} = 0$. This reflects the fact that the quasiparticle distribution $f_{\rm F+}$ created by the tunneling of energy-imbalanced quasiparticles can contribute to the tunneling current only when the energy relaxation time for $f_{\rm F+}(y,\epsilon)$ is different from that for $f_{\rm F+}(y,-\epsilon)$.[20] That is, the energy relaxation process is essential in obtaining a nonzero $I_{\rm F}$.

We obtain $I_{\rm F}$ and $I_{\rm S}$ by substituting the resulting quasiparticle distributions into eqs. (15) and (28). The three terms are given as follows:

$$I_{\rm q} = \chi(T)\frac{V_{\rm det}}{R_{\rm det}}, \tag{45}$$

$$I_{\rm F} = \frac{3R_{\rm S}R_{\rm F}}{8R_{\rm inj}R_{\rm det}} \frac{L\lambda_{\rm e}(\Delta)}{\lambda_{\rm sf}^2} J_3(V_{\rm inj},T)\frac{V_{\rm det}}{R_{\rm det}}, \tag{46}$$

$$I_{\rm S} = \eta\frac{R_{\rm S}}{2R_{\rm inj}} P_{\rm spin}^2 {\rm e}^{-\frac{d}{\lambda_{\rm sf}}} J_2(V_{\rm inj},T)\frac{\Delta}{eR_{\rm det}}, \tag{47}$$

where $\eta = 1(-1)$ for the parallel (antiparallel) alignment and

$$\chi(T) = \int_0^\infty {\rm d}\epsilon N_1(\epsilon)\left(-2\frac{\partial f_0(\epsilon)}{\partial\epsilon}\right), \tag{48}$$

$$J_3(V,T) = \frac{1}{\Delta}\int_0^\infty {\rm d}\epsilon N_1^3(\epsilon)\big(f_0(\epsilon - eV) + f_0(\epsilon + eV) - 2f_0(\epsilon)\big), \tag{49}$$

$$J_2(V,T) = \frac{1}{\Delta}\int_0^\infty {\rm d}\epsilon N_1^2(\epsilon)\big(f_0(\epsilon - eV) - f_0(\epsilon + eV)\big). \tag{50}$$

The resistances $R_{\rm S}$ and $R_{\rm F}$ are defined by $R_{\rm S} \equiv \lambda_{\rm sf}\rho_{\rm S}/A_{\rm S}$ and $R_{\rm F} \equiv \lambda_{\rm sf}\rho_{\rm F}/A_{\rm F}$ with the resistivities $\rho_{\rm S} = (2e^2 N_{\rm S} D_{\rm S})^{-1}$ and $\rho_{\rm F} = (2e^2 N_{\rm F} D_{\rm F})^{-1}$. It should be noted that $J_3(V,T)$ and $J_2(V,T)$ diverge if eq. (35) is adopted as the expression of $N_1(\epsilon)$. This unphysical divergence does not arise if we adopt a more realistic expression of $N_1(\epsilon)$, which does not diverges at the gap edge. Indeed, the divergence of $N_1(\epsilon)$ is actually removed if we take account of gap anisotropy, inelastic electron scattering or magnetic impurity scattering. We obtain $V_{\rm p}$ and $V_{\rm ap}$ by solving $I_{\rm det}(V_{\rm p}) = 0$ for $\eta = 1$ and $I_{\rm det}(V_{\rm ap}) = 0$ for $\eta = -1$, respectively. Substituting the resulting expressions and eq. (10) into eq. (1), we finally obtain

$$R_{\rm spin} = \gamma P_{\rm spin}^2 R_{\rm S} {\rm e}^{-\frac{d}{\lambda_{\rm sf}}} \tag{51}$$

with

$$\gamma = \frac{J_2(V_{\rm inj},T)}{J_1(V_{\rm inj},T)\left(\chi(T) + \frac{3R_{\rm S}R_{\rm F}}{8R_{\rm inj}R_{\rm det}}\frac{L\lambda_{\rm e}(\Delta)}{\lambda_{\rm sf}^2}J_3(V_{\rm inj},T)\right)}. \tag{52}$$

Note that γ represents the renormalization of the spin signal induced by the transition to the superconducting state, and $\gamma = 1$ corresponds to the normal state.

In the remaining of this section, we briefly consider the influence of magnetic impurities. We redefine $\tau_{\rm sf}$ as

$$\frac{1}{\tau_{\rm sf}} = \frac{4}{3\tau_{\rm so}} + \frac{4}{3\tau_{\rm m}}, \tag{53}$$

and introduce the parameter[11]

$$\beta = \frac{\tau_{\rm so} - \tau_{\rm m}}{\tau_{\rm so} + \tau_{\rm m}} \tag{54}$$

which characterizes the relative strength of spin-orbit scattering and magnetic impurity scattering. Here, $\tau_{\rm sf}$ should be regarded as the spin-flip scattering time in the normal state. We observe that $\beta = -1$ in the absence of magnetic impurities and $\beta = 1$ when spin-orbit scattering does not occur. If $\beta \neq -1$, we must solve eq. (16) to obtain the spectral functions. Strictly speaking, eqs. (35)

and (36) are not justified in the presence of magnetic impurities and the relation $N_1^2(\epsilon) - R_2^2(\epsilon) = 1$ no longer holds exactly. Consequently, $f_{L+}(x_{det}, \epsilon)$ and $f_{L-}(x_{det}, \epsilon)$ are modified as

$$f_{L+}(x_{det}, \epsilon) = \frac{P_{spin}\alpha(\epsilon)\lambda_{sf}}{8e^2 N_S A_S R_{inj} D_S} \frac{\Sigma_+(\epsilon, V_{inj})}{(N_1^2(\epsilon) - R_2^2(\epsilon))} \times e^{-\frac{d}{\alpha(\epsilon)\lambda_{sf}}}, \qquad (55)$$

$$f_{L-}(x_{det}, \epsilon) = \frac{L}{8e^2 N_S A_S R_{inj} D_S} \frac{\Sigma_-(\epsilon, V_{inj})}{(N_1^2(\epsilon) - R_2^2(\epsilon))}, \qquad (56)$$

where

$$\alpha(\epsilon) = \sqrt{\frac{N_1^2(\epsilon) - R_2^2(\epsilon)}{N_1^2(\epsilon) + \beta R_2^2(\epsilon)}}. \qquad (57)$$

The parameter $\alpha(\epsilon)$ represents the renormalization of the spin-flip scattering time on transition to the superconducting state.[15] Using eqs. (55) and (56), we can show that eqs. (51) and (52) are applicable to this case if J_3 and J_2 are replaced by the following expressions,

$$J_3(V, T) = \frac{1}{\Delta} \int_0^\infty d\epsilon \frac{N_1^3(\epsilon)}{N_1^2(\epsilon) - R_2^2(\epsilon)} \big(f_0(\epsilon - eV) + f_0(\epsilon + eV) - 2f_0(\epsilon) \big), \qquad (58)$$

$$J_2(V, T) = \frac{1}{\Delta} \int_0^\infty d\epsilon \frac{\alpha(\epsilon) N_1^2(\epsilon)}{N_1^2(\epsilon) - R_2^2(\epsilon)} e^{-(\alpha(\epsilon)^{-1} - 1)\frac{d}{\lambda_{sf}}} \times \big(f_0(\epsilon - eV) - f_0(\epsilon + eV) \big). \qquad (59)$$

We here comment on the expression of the spin signal presented by Poli *et al.*[11] We note that they ignore the influence of nonequilibrium quasiparticles in the ferromagnetic metal for detection and therefore the corresponding term is lacking. This is the significant difference between their expression and ours. In addition, they assume $V_{inj} \ll \Delta/e$. Finally, we point out that $\alpha(\epsilon)$-dependence is slightly different between them. Indeed, if the factor $2\alpha + N(E)R_N/R_I$ in eq. (4) of ref. 11 is replaced by $2\alpha^{-1}$, their expression becomes nearly identical to ours in the case of $J_3(V_{inj}, T) = 0$ and $V_{inj} \ll \Delta/e$. The reason for this difference is not clear.

4. Discussion

Let us consider the temperature dependence of the renormalization factor γ under the condition that the injection current I_{inj} is kept constant. We adjust V_{inj} to supply a constant injection current. This means that V_{inj} is determined as a function of T for a given I_{inj}, so we rewrite $J_i(V_{inj}, T)$ as $J_i(I_{inj}, T)$ $(i = 1, 2, 3)$. It should be noted here that even though I_{inj} is very small, V_{inj} approaches to Δ/e as $T \to 0$.

We focus on the low temperature regime where the T-dependence of Δ can be neglected. In this regime, $\chi(T)$ behaves as

$$\chi(T) = \sqrt{\frac{2\pi\Delta}{T}} e^{-\frac{\Delta}{T}}. \qquad (60)$$

When T is not very low and $\chi(T)$ is much greater than the term with $J_3(I_{inj}, T)$ in the denominator of eq. (52), the renormalization factor is reduced to

$$\gamma = \frac{J_2(I_{inj}, T)}{J_1(I_{inj}, T)\chi(T)}. \qquad (61)$$

Because the T-dependence of $J_2(I_{inj}, T)/J_1(I_{inj}, T)$ is weak, we obtain $\gamma \propto \chi(T)^{-1}$. This indicates that γ behaves as $\gamma \propto e^{\Delta/T}$. However, because $\chi(T)$ is exponentially suppressed with decreasing T, the term with $J_3(I_{inj}, T)$ eventually dominates $\chi(T)$ below a crossover temperature T_{cross}. Below T_{cross}, we can ignore $\chi(T)$ in eq. (52) and the renormalization factor is reduced to

$$\gamma = \frac{8R_{inj}R_{det}}{3R_S R_F} \frac{\lambda_{sf}^2}{L\lambda_e(\Delta)} \frac{J_2(I_{inj}, T)J_3(I_{inj}, T)}{J_1(I_{inj}, T)}. \qquad (62)$$

The crossover temperature is determined by

$$\chi(T_{cross}) = \frac{3R_S R_F}{8R_{inj}R_{det}} \frac{L\lambda_e(\Delta)}{\lambda_{sf}^2} J_3(I_{inj}, T_{cross}). \qquad (63)$$

As T is lowered below T_{cross}, the injection voltage V_{inj} approaches to Δ/e. In this situation, the T-dependence of $J_3(I_{inj}, T)$ becomes weak. Furthermore, we can neglect the weak T-dependence of $J_2(I_{inj}, T)/J_1(I_{inj}, T)$. Thus, we conclude that below T_{cross}, the renormalization factor γ rapidly converges to the value given by $\gamma_0 \equiv \lim_{T\to 0} \gamma$. We can obtain γ_0 from eq. (62) with $T = 0$.

From the above argument, we observe the qualitative behavior of R_{spin} as follows. In the regime of $T \gg T_{cross}$, the spin signal exponentially increases with decreasing T as $R_{spin} \propto e^{\Delta/T}$. Below T_{cross}, however, the spin signal converges as $R_{spin} \to \gamma_0 P_{spin}^2 R_S e^{-d/\lambda_{sf}}$. We here point out that the behavior of R_{spin} in the regime of $T \gg T_{cross}$ is qualitatively equivalent to the previous result, eq. (3), reported by Takahashi and Maekawa.[5] However, our argument indicates that the exponential increase of R_{spin} should not be attributed to the increase of spin resistivity.[5] We simply understand that R_{spin} increases reflecting the suppression of thermally excited quasiparticles in the detection junction.

Let us consider the experimental result reported by Poli *et al.*[11] on the basis of our theoretical framework. Particularly, we focus on the convergence of R_{spin} observed at low temperatures. They employed the device consisting of a superconducting wire of Al and ferromagnetic metal wires of Co. Since it has been believed that spin-flip scattering in Al is mainly caused by spin-orbit interaction, we assume that magnetic impurity scattering is much less relevant than spin-orbit scattering and set $\tau_m^{-1} = 0$. We estimate the limiting value γ_0 of the renormalization factor from eq. (62) with $T = 0$ and compare it with their experimental value. Following refs. 10 and 11, we employ the parameters: $I_{inj} = 1$ nA, $\lambda_{sf} = 1$ μm, $\Delta = 200$ μeV, $R_{inj} = R_{det} = 100$ kΩ, $\rho_S = 10$ $\mu\Omega$cm, $A_S = 10 \times 150$ nm^2, $A_F = 50 \times 130$ nm^2. For the other parameters, we assume $L = 10$ μm, $D_F = 3 \times 10^{-3}$ m^{2}s^{-1}, $\rho_F = 14$ $\mu\Omega$cm, $\alpha_F = 9 \times 10^3$ eV^{-2}. The value of α_F is estimated by using the relation[18] $\alpha_F \sim 2/\tau_D T_D^3$ with $T_D = 385$ K and $\tau_D = 0.4 \times 10^{-14}$ s, where T_D and τ_D are the Debye temperature and the phonon scattering time at T_D, respectively. From these parameters, we obtain $\lambda_e(\Delta) = 9$ μm, $R_S = 67$ Ω and $R_F = 22$ Ω. The integral $J_1(I_{inj}, T)$ does not depend on T and is obtained from

eq. (10) as $J_1(I_{\rm inj}, T) = eR_{\rm inj}I_{\rm inj}/\Delta = 0.5$. We finally consider $J_2(I_{\rm inj}, T)$ and $J_3(I_{\rm inj}, T)$ in the limit of $T \to 0$. The evaluation of these integrals is not simple, so we roughly approximate them as $J_2(I_{\rm inj}, 0) = J_3(I_{\rm inj}, 0) = J_1(I_{\rm inj}, T)$. Substituting these parameters into eq. (62), we approximately obtain $\gamma_0 \sim 10^5$. This indicates that $R_{\rm spin}$ below $T_{\rm cross}$ is a factor of 10^5 larger than that in the normal state. This is consistent with the experimental result which indicates the enhancement of 4 or 5 orders of magnitude. We estimate the crossover temperature by solving eq. (63) with eq. (60) and obtain $T_{\rm cross} \sim 0.1$ K. This is also consistent with the experimental value of $T_{\rm cross} \sim 0.16$ K.

We have shown that nonequilibrium quasiparticles with $|\epsilon| \approx \Delta$ are created in the ferromagnetic metal for detection by the tunneling of energy-imbalanced quasiparticles, and that these quasiparticles contribute to $I_{\rm det}$ in combination with the energy relaxation process due to phonon scattering. It should be noted that the energy relaxation of quasiparticles excites phonons near the detection junction, leading to the increase of effective temperature $T_{\rm eff}$ for quasiparticles. If $T_{\rm eff}$ becomes greater than $T_{\rm cross}$, the convergence of the spin signal is determined by this heating effect instead of the convergence mechanism which we discussed above. The separation of these two mechanisms is a future problem for experiments.

In addition to the heating effect, we have ignored charge imbalance. If the injection current is not small, we must consider its influences. Charge imbalance provides a nearly constant contribution $I_{\rm Q}$ to $I_{\rm S}$ regardless of the alignment of magnetizations. Since $I_{\rm Q}$ must be cancelled by $I_{\rm q}$ and $I_{\rm F}$ to ensure $I_{\rm det} = 0$, we expect that both $V_{\rm p}$ and $V_{\rm ap}$ increases with increasing $I_{\rm Q}$. However, if $I_{\rm Q}$ is sufficiently small, the increase of $V_{\rm p}$ is equivalent to that of $V_{\rm ap}$ because both $I_{\rm q}(V_{\rm det})$ and $I_{\rm F}(V_{\rm det})$ linearly depends on $V_{\rm det}$ when $|V_{\rm det}| \ll \Delta/e$. Therefore, we expect that no qualitative change of the spin signal appears as long as charge imbalance is not very large.

In summary, we have studied the transport of spin-polarized nonequilibrium quasiparticles in a superconducting wire connected by tunnel junctions to two ferromagnetic metal wires, each of which serves as a spin injector or detector. We have presented a basic formalism to determine spin-polarized quasiparticle distributions in this system, and obtained an analytical expression for the nonlocal spin signal. We have taken account of nonequilibrium quasiparticles in the ferromagnetic metal for detection, which are created by the tunneling of energy-imbalanced quasiparticles in the superconductor. We have shown that they induce the convergence of the spin signal at low temperatures.

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